

Aerosol processing in coastal stratocumulus clouds



Mikhail Ovtchinnikov

Richard Easter



Pacific Northwest
National Laboratory
Operated by Battelle for the
U.S. Department of Energy

Richland, WA, USA

1. INTRODUCTION

MOTIVATION:

Aerosol properties are affected by clouds with implications for direct and indirect aerosol effects. The physics of aerosol-cloud interaction is complex and includes:

- transport (*advection, sedimentation, mixing*);
- cloud microphysics (*in- and below-cloud scavenging, coalescence, resuspension*);
- aqueous chemistry.

Many of these processes are crudely represented in regional and global models, and the effects of clouds on AP size distribution is poorly understood. Accuracy and applicability of many *ad hoc* assumptions and approximations is not known.

CHALLENGE: To track aerosol transformations inside cloud droplets

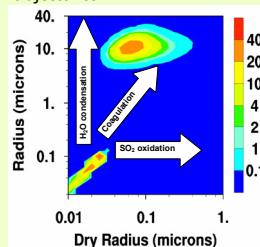
APPROACH:

To model evolution of aerosol particles (APs) and cloud droplets using a joint two-dimensional size distribution, in which the wet particle size/volume and the dry particle size/volume are two independent variables. This formulation preserves the dry distribution of APs exactly during the water condensation-evaporation cycle and provides a framework for predicting changes in the AP distribution due to drop coalescence and aqueous chemistry.

The new microphysics module has been implemented in a fully interactive multidimensional LES framework and a parcel model driven by an ensemble of trajectories.

Fig. 1. A diagram illustrating how the 2D distribution function is affected by different processes. Only part of the size domain is shown. Full domain consists of 1200 bins of the two-dimensional particle distribution:

30 dry size categories over a range of radii 0.01 – 10 μm ; 50 wet size categories over a range of 0.02 – 1.65 μm . Currently, a uniform chemical AP composition is assumed (ammonium sulfate in this study), and limited aqueous chemistry is simulated.



2. MASE 25 JULY 2005 CASE STUDY

LES model is driven by prescribed time-dependent ocean temperature and vertical profiles of temperature, moisture, and horizontal wind. The model domain covers $3 \times 3 \times 0.9 \text{ km}^3$ with $50 \times 50 \times 60$ grid cells at a resolution $60 \times 60 \times 15 \text{ m}$. The base simulation is run for three hours without aqueous chemistry. In another run, the aqueous chemistry is turned on after an hour of spin up.

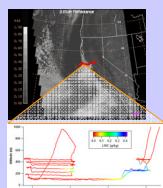


Fig. 2. Satellite view of the cloud field with superimposed G-1 flight track (upper panel). The lower panel shows the height-longitude projection of the flight track with color-coded liquid water content.

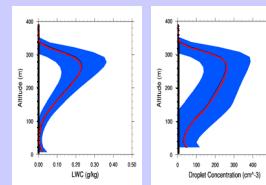


Fig. 3. Vertical profiles of liquid water content (left) and droplet number concentration (right), mean plus and minus one standard deviation.

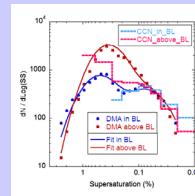


Fig. 4. The DMA-measured (dots) and fitted (solid lines) size distributions converted to CCN supersaturation spectra assuming ammonium sulfate composition are in good agreement with CCN measurements (dashed lines) in the boundary layer but not above.

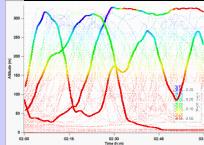


Fig. 5. A spaghetti diagram of 50 out of 500 trajectories generated by the LES model and used in the ensemble parcel model simulations. Three drastically different trajectories are highlighted.

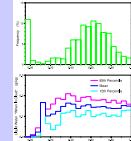


Fig. 6. Cloud statistics of all 500 trajectories.

3. RESULTS

The case-study clouds have low liquid water content and high droplet concentration, so the droplet collision efficiencies are small and coagulation has been neglected. Changes in the aerosol spectrum are driven by aqueous-phase oxidation of SO_2 by H_2O_2 and O_3 . All simulations show shifts of aerosol number concentration from Aitken to accumulation mode but the rate of this transfer depends strongly on aerosol composition and concentrations of gases.

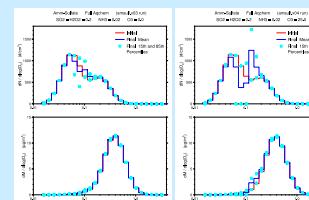


Fig. 7. Initial (red) and processed (blue) dry size distribution of aerosol from LES (top) and ensemble trajectory (bottom) models when only aqueous-phase oxidation of SO_2 by H_2O_2 accounted for.

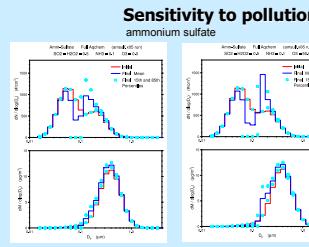


Fig. 8. Ensemble trajectory model results with and without $\text{SO}_2 + \text{O}_3$ reaction. Accounting for oxidation by ozone significantly increases the aerosol processing rate for a non-acidic ammonium ammonium sulfate aerosol. The smallest activated particles have largest changes because they contain relatively more water (higher water/salt ratio)

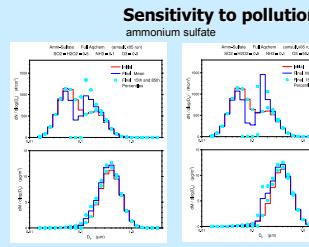


Fig. 9. Ensemble trajectory model results with higher SO_2 and oxidant levels show larger changes to the aerosol distributions, and a greater contribution from $\text{SO}_2 + \text{O}_3$ for ammonium sulfate aerosol than for acidic ammonium bisulfate aerosol.

4. DISCUSSION

A 2D (dry size / wet size) representation of the atmospheric particle spectrum is a useful and powerful although expensive tool to study aerosol – cloud interactions. The module is integrated into two frameworks that compliment each other.

Ensemble-Trajectory Lagrangian Parcel Model

Benefits:

- Relatively inexpensive to run;
- Option 1 (one or more 1D size distributions with moving bins) allows treating external mixtures and complex, drop-size dependent aqueous chemistry;
- Option 2 (2D size distribution) allows for adding and testing other multi-moment distribution function (e.g., number, water mass, and AP chemical component masses in each bin).

Limitations:

- Lacking feedback between microphysics and dynamics;
- A closed parcel assumption: mixing and sedimentation neglected or highly simplified (or parameterized).

Fully Interactive LES Model

Benefits:

- Allows for feedbacks between microphysics and dynamics;
- Explicit treatment of mixing and sedimentation.

Limitations:

- Expensive to run (remember ~ 1200 variables per grid cell);
- Accuracy (because of coarser bin resolution);
- One-moment scheme for microphysics* (only particle number concentration in each 2D bin);
- Single composition aerosol* (e.g., ammonium sulfate).

*Two moment scheme with 2-3 aerosol chemical components may be possible in the future.

Our ultimate objective is to use results from detailed simulations of cloud processing of AP to develop/evaluate parameterizations for use in global models that have simpler treatments of both cloud microphysics (e.g., bulk) and aerosols (e.g., modal).

Acknowledgments

This research is supported by the DOE Atmospheric Science Program.



Special thanks to Stephen Springston, Jim Hudson, and John Huber for providing and guiding us through the data; Chris Waleck for advise and the advection scheme; and PNNL Advanced Computing Center for providing computing resources.